

Chapter 1: Increased Use of Ethanol in Gasoline and Potential Ground Water Impacts

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1. Introduction: Increased Use of Ethanol in Gasoline and Potential Ground Water Impacts

1.1. Background

1.1.1. MtBE Phase-out

Over the past five years, a significant debate has developed surrounding the use of oxygen-bearing chemicals as additives for automotive fuels (Franklin *et al.*, 2000, Nadim *et al.*, 2001). The increase in the use of methyl tertiary butyl ether (MtBE) from a minor gasoline component to 11% by volume to meet the requirements of the Clean Air Act Amendments of 1990 has resulted in widespread contamination of ground and surface waters and has adversely affected drinking water supplies (Zogorski *et al.*, 1996; Gullick and LeChevallier, 2000; Johnson *et al.*, 2000). The recognition that regulatory decisions to improve the quality of one environmental medium could be so detrimental to other environmental media has resulted in the hopes that future regulations would carefully consider the broad impacts of such changes prior to implementation (Franklin *et al.*, 2000; Davis and Farland, 2001). It is clear that the environmental, economic, and societal impacts need to be better understood in order to make informed regulatory decisions regarding the composition of our nation's gasoline.

The phase-out of MtBE as a gasoline additive in California and several other states has placed renewed interest in the use of ethanol as a gasoline oxygenate. Ethanol can be used in oxygenated gasoline ~8% in oxyfuel or ~6% in reformulated gasoline (RFG), by volume. It is also sometimes used at 10% volume in gasoline, even in areas that are not required to use RFG, because there is a federal subsidy to promote markets for fuel-grade ethanol (RFA, 1999). Gasoline with ethanol added as an oxygenate at 10% or less by volume is generally referred to as "gasohol."

At the federal level, several legislative bills that could affect the use of ethanol in gasoline are presently being debated within our federal government (Table 1-1). The congressional action regarding ethanol includes its value as a gasoline additive because it is considered a renewable fuel as well as an oxygen-bearing chemical. While debate continues over these bills, it is critical that research be conducted to help understand—to the greatest extent practical—the overall impacts that would be caused by the implementation of these bills.

1.1.2. Use of Ethanol-blended Gasoline

Worldwide, the most significant use of ethanol started in Brazil in the 1970s. In that country, the National Alcohol Program was created to cope with the high oil prices of the 70s and 80s. Federal incentives, in combination with the participation of the automobile industry and the strong environmental appeal, made the program a success. In 1985, 95% of all cars produced used hydrated ethanol as the only fuel. When oil prices began to drop, the program was changed because the sale of cars that could run only on ethanol plummeted. In 1999, approximately 75% of all automobiles in Brazil ran on gasoline containing 24% ethanol, with a total fuel alcohol consumption of 13.8×10^6 m³/year (ANP, 1999).

Table 1-1. Some of the current legislation that could result in an increase in the use of fuel-grade ethanol (adapted from material presented by the Governor's Ethanol Coalition, 2001).

Bill No.	Sponsor	Title	Description
S.265	Illinois Senator Fitzgerald	MtBE Elimination Act	Bans MtBE
S.670	South Dakota Senator Daschle	Renewable Fuels Act of 2001	Bans MtBE; creates a Renewable Fuel Requirement; maintains oxygenate requirement in RFG
S.892	Iowa Senator Harkin	Clean & Renewable Fuels Act of 2001	Bans MtBE; creates a Renewable Fuel Requirement; maintains oxygenate requirement in RFG
S.950	New Hampshire Senator Smith	Federal Reformulated Fuels Act of 2001	Bans MtBE; eliminates oxygenate requirement in RFG; eliminates volatility waiver for ethanol blends in conventional gasoline; provides funding for stranded investments by MtBE producers
S.1006	Nebraska Senator Hagel	Renewable Fuels for Energy Security Act of 2001	Creates a Renewable Fuel Requirement for gasoline and diesel.
H.R.2017	Wisconsin Representative Green		Directs EPA to conduct a study of feasibility of developing regional fuel specifications, including a uniform Midwestern fuel that includes ethanol
H.R.2249	Missouri Representative Blunt	Gasoline Access & Stabilization Act of 2001	Creates a more uniform formula for gasoline by reducing the number of gasolines to conventional gasoline, oxygenated reformulated gasoline and California gasoline.
H.R.2303	Kentucky Representative Lewis	Ethanol and Biodiesel Promotion Act of 2001	Would amend the internal revenue code to provide incentives to increase sales of ethanol and biodiesel fuel blends

In the United States, the push to use ethanol as a biomass fuel has resulted in its use in several states, particularly in the Midwest. Gasohol containing 10% ethanol by volume has been used extensively in the corn-growing states of the Midwest for several years. It can be found as an option at most gasoline stations in Iowa and Nebraska, accounting for 21% of all motor fuel sold in Nebraska (Chambers, 1999). In 1999, 60% of gasoline sold in Illinois and 90% of gasoline sold in the Chicago area contained 10% ethanol (RFA, 1999). Throughout the country, U.S. consumers used more than 56 million cubic meters (15 billion gallons) of ethanol-blended gasoline in 1999 (ACE, 1999)

Governor Gray Davis issued Executive Order D-5-99 on March 25, 1999, calling for the removal of MtBE from gasoline at the earliest possible date but no later than December 31, 2002. California also asked the U.S. EPA to grant a waiver of the mandate to use oxygenates in California gasoline. This request for an oxygenate waiver has been denied.

The phase-out of MtBE in several states, coupled with the U.S. EPA decision to deny California's request for a waiver from the oxygenate requirement in federal RFG, has the potential to greatly increase the use of ethanol as a component in gasoline in this country. It has been estimated that California will use approximately 150 million gallons of ethanol during 2001 (Schremp, 2001). After the MtBE phase-out deadline of December 31, 2002, California's ethanol demand is expected to increase to 660 to 950 million gallons per year (Schremp, 2001).

1.2. Areas of Subsurface Fate and Transport Uncertainty

Task 10 of Executive Order D-5-99 states "the California Air Resources Board (ARB) and the State Water Resources Control Board (SWRCB) shall conduct an environmental fate and transport analysis of ethanol in air, surface water, and groundwater. The Office of Environmental Health Hazard Assessment (OEHHA) shall prepare an analysis of the health risks of ethanol in gasoline, the products of incomplete combustion of ethanol in gasoline, and any resulting secondary transformation products." In December 1999, a report entitled *Health and Environmental Assessment of the Use of Ethanol as a Fuel Oxygenate* (Rice and Cannon [Eds.], 1999) summarized the results of a screening analysis performed in fulfillment of the requirements of the Executive Order. The screening analysis also identified several uncertainties associated with the potential impacts to ground and surface water from increased use of ethanol in gasoline. These results were presented to the California Environmental Policy Council in January of 2000.

As a result of this report, the California Environmental Policy Council found that from an environmental management standpoint, the impacts associated with the use of ethanol will be significantly less as well as more manageable than those associated with the continued use of MtBE, but that further research was warranted.

An important research consideration is the effect ethanol in gasoline may have on the subsurface fate and transport of toxic gasoline components, in particular, the monoaromatic components, benzene, toluene, ethylbenzene, and xylenes (BTEX). This fate and transport information is important for evaluating the impact that ethanol may have on the cleanup of gasoline releases and on California's water resources in general.

The hygroscopic nature of gasohol prevents its preparation at a refinery and distribution by pipeline. Thus, gasoline distribution terminals receive gasoline and ethanol separately to be mixed and pumped into the tanker truck for delivery to a gasoline station (Figure 1-1). It is presently anticipated that most of the ethanol used in California would be produced in the Midwest, denatured with gasoline (5%) and shipped to California by rail (30,000-gallon rail cars or by marine cargo (shipments of millions of gallons), followed by rail or truck delivery to a final destination terminal.

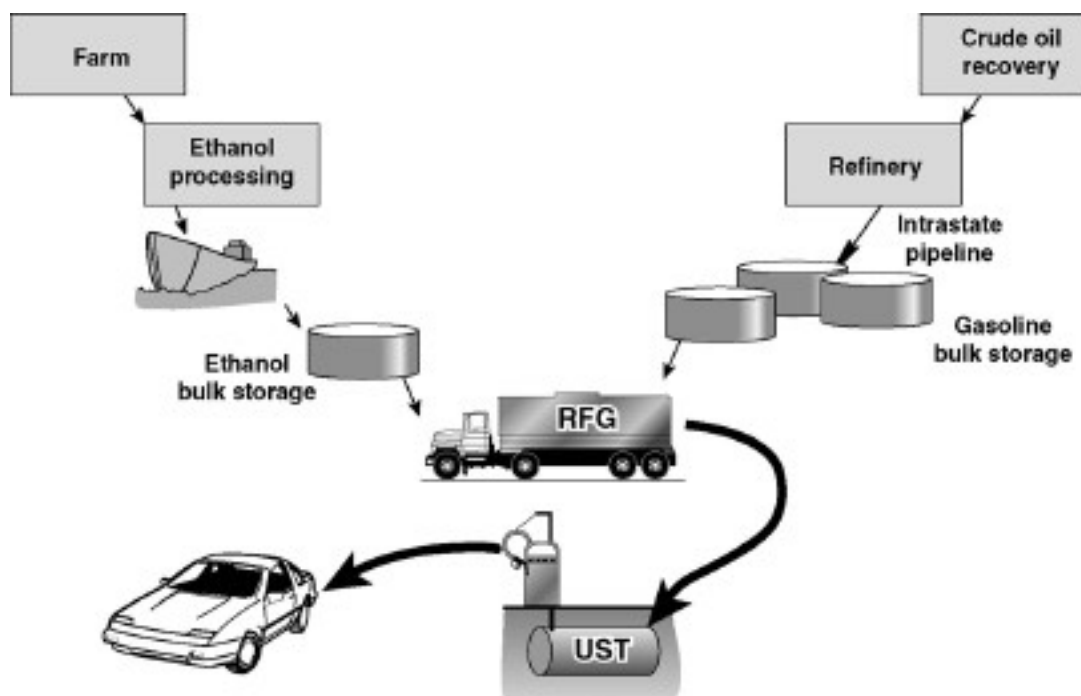


Figure 1-1. Life cycle of ethanol-blended gasoline.

Given the manner in which gasohol is mixed and distributed, there are several potential scenarios for spills of ethanol or gasohol gasoline to the subsurface. Rice et al. (1999) summarized the potential for different spill events.

The events most likely to affect soil and the subsurface include:

- Release at a gasoline station during the filling of an underground storage tank (UST) (< 50 gallons)
- Release (potentially undetected) at a gasoline station from a small puncture in an UST (< ~3 gal/d)

The events moderately likely to affect soil and the subsurface include:

- Release of bulk ethanol at a distribution terminal (> 150,000 gallons)
- Release of gasohol during transport by tanker truck to a gasoline station (~ 5000 gallons)
- Release at a gasoline station from a large puncture in an UST (< ~10 gal/d)

Two very different scenarios are included above: (1) spills of gasohol and (2) spills of bulk denatured ethanol, most likely into soil that has already been contaminated by fuel hydrocarbons. It is expected that the concentration of ethanol in the soil and groundwater would vary greatly depending on which scenario would apply. As described throughout this report, this would lead

to significant differences in the behavior of petroleum hydrocarbons in association with the ethanol.

Several abiotic processes that affect the fate of ethanol and ethanol-gasoline mixtures in the subsurface have been identified. These processes include infiltration, spreading at the capillary fringe, and leaching of chemicals into groundwater. These abiotic processes can potentially impact the retention and distribution of gasohol or other petroleum products in the unsaturated zone, the size and the shape of a gasoline pool at the water table, and the flux of contaminants from the gasoline to the groundwater.

In addition to abiotic processes, biodegradation of fuel ethanol contributes to the depletion of electron-acceptors in groundwater that are needed to biodegrade other gasoline components, such as BTEX compounds. This depletion is important to study because it may affect both BTEX degradation and migration rates. For example, both the changes in electron acceptor availability and the presence of easily degradable ethanol could affect populations and metabolic activities of indigenous hydrocarbon-degrading bacteria.

1.2.1. Unsaturated Zone Transport

We have very little information on the behavior of gasohol as it infiltrates through the unsaturated zone. On the other hand, there is substantial knowledge about many of the mechanisms affecting saturated zone transport of gasohol. The net effect of ethanol on the length and longevity of a contaminant plume, however, requires an understanding of each of the steps that define the complete transport pathway. Rather than just deal with each of these steps individually, it is important to understand the complex interrelationships among the processes involved with the ultimate transport of gasoline components to a potential downgradient receptor. Understanding the processes affecting the migration of gasoline components through the unsaturated zone in the presence of ethanol is crucial because knowledge gaps about the early stages of the overall flow and transport preclude adequate prediction of the impacts of ethanol on BTEX contamination. To better understand these unsaturated zone processes, additional research was conducted. The results of this research are presented in Chapter 2 of this report, *Infiltration and Distribution of Ethanol and Ethanol-blended Gasolines in the Vadose Zone*.

1.2.2. Impact of Ethanol on Benzene, Toluene, Ethylbenzene, and Xylene Groundwater Plumes

Several modeling studies have predicted that, in the presence of ethanol, benzene plumes are likely to increase in length. The amount of this increase is not well known.

These groundwater modeling studies have included several simplifying assumptions, including the following: (1) there will be preferential biodegradation of dissolved ethanol near the release source area, (2) no benzene degradation will occur within this ethanol degradation zone, (3) downgradient of the ethanol degradation zone, there will be a depletion of available electron acceptors that will result in lower benzene degradation rates, and (4) the benzene degradation rate is constant in time and space. These assumptions, if incorrect, would tend to overestimate predicted benzene plume lengths.

Since microbially-mediated processes appear to dominate the fate and transport of gasoline components in the presence of ethanol, additional research was conducted to gain a better

understanding of the impact ethanol may have on BTEX biodegradation kinetics. The results of this research are presented in Chapter 3 of this report, *Effect of Ethanol and MtBE on BTEX Biodegradation in the Saturated Zone: Kinetic Studies*.

Research was also conducted to better understand how ethanol can affect the composition of subsurface microbial communities, with an emphasis on indigenous bacterial populations capable of BTEX degradation. This represented an effort to better understand ecological change underlying the effects of ethanol on BTEX degradation kinetics that are reported in Chapter 3 of this report. The results of this research are presented in Chapter 4 of this report, *Effect of Ethanol on Hydrocarbon-degrading Bacteria in the Saturated Zone: Microbial Ecology Studies*.

A more sophisticated modeling approach that better represents the spatial and temporal transport of electron acceptors is presented in Chapter 5 of this report, *A Finite-difference-based Reactive Transport Model Assessment of the Effects of Ethanol Biotransformation on the Lengths of Benzene Plumes from Leaking Underground Fuel Tanks*.

1.2.3. Evaluation of Ethanol Sampling and Analytical Methods

Improved routine sampling and analysis methods are needed to meet the data quality objectives of future studies and groundwater resource management. A study was performed to evaluate the best storage and contract laboratory analytical protocols for environmental water samples containing ethanol and to make recommendations for improvement. The results of this evaluation are reported in Chapter 6 of this report, *Evaluation of Storage and Analysis Protocols for Environmental Water Samples Containing Ethanol*.

1.3. Potential Effects of Ethanol on Site Remediation Activities

Several physicochemical and biological processes can be used to treat BTEX-contaminated aquifers. Early approaches relied primarily on pumping the contaminated groundwater for above-ground treatment with activated carbon or air strippers. Nevertheless, BTEX compounds are moderately hydrophobic and tend to sorb to the aquifer material, which makes them difficult to withdraw by pumping. Thus, pump-and-treat alone can result in prohibitively long time periods for the removal of the residual contamination, and is often economically unfeasible (Environmental Engineering Research Council, 1990). The ubiquity of aerobic microorganisms capable of degrading BTEX has been established, and many state-of-the-art remediation approaches rely on the exploitation of such natural degradative processes *in situ*. The most common of these bioremediation processes are discussed below, followed by a brief prognosis of how the presence of ethanol may affect process performance.

1.3.1. BTEX Mass Extraction Approaches

Mass extraction techniques have been used extensively to clean up gasoline released to the subsurface. The process involves drilling extraction wells that are screened either above or below the water table or both. The most common mass extraction approaches to treat BTEX contamination are (National Research Council, 1994):

- **Soil Vapor Extraction:** This method flushes air through soil above the water table to extract volatile gasoline components in the unsaturated zone. This process is limited by the permeability of the soils and the degree to which the fuel hydrocarbons are bound to

the soils. The extracted vapors are typically treated at the surface using either direct combustion or sorption onto granulated activated carbon.

- **Ground Water Pump and Treat:** This approach involves the pumping of large volumes of groundwater to capture dissolved BTEX plumes and mobilize and extract dissolved fuel components. The process is limited by the physical heterogeneity of the subsurface which allows preferential pathways that bypass trapped contaminated water, the slow dissolution of fuel hydrocarbons into the groundwater, and sorption of fuel hydrocarbons on the soils. The extracted groundwater typically is treated at the surface using a variety of processes including air stripping, sorption onto granulated activated carbon, or chemical destruction.

1.3.2. BTEX Bioremediation Approaches

Bioremediation is a managed or spontaneous process in which biological, especially microbiological, catalysis acts on pollutants, thereby remedying or eliminating environmental contamination. The most common bioremediation approaches to treat BTEX contamination are (Rittmann and McCarty, 2001):

- **Bioventing:** This approach is used to stimulate aerobic degradation processes above the water table by the action of vacuum pumps that pull air through the unsaturated zone. Bioventing is often used with infiltration galleries that deliver water (with nutrients) to prevent desiccation in the unsaturated zone.
- **Water Circulation Systems:** This method is based on extracting contaminated groundwater for above-ground treatment and reinjecting it into the ground with stimulatory amendments (e.g., H_2O_2 as an oxygen source, and nutrients). Clogging near injection well screens and infiltration galleries can occur due to bacterial growth and mineral precipitation. In general, pulsing nutrients results in less clogging than continuous delivery. Occasional pulsing of Cl_2 to control biofouling can also prevent clogging.
- **Air Sparging:** This engineered system involves injection of compressed air into the contaminated subsurface to deliver oxygen and strip the BTEX into a vapor-capture system. Air sparging can be a relatively effective and inexpensive BTEX bioremediation approach, but it is not effective when low-permeability soil traps or diverts the airflow.
- **Biobarriers:** This term refers to biologically active zones that are placed in the path of narrow BTEX plumes, often incorporating air spargers (i.e., air-curtains) or oxygen-releasing compounds to enhance oxidative biodegradation processes. Hydraulic or physical controls on groundwater movement may be required to ensure that BTEX pass through the barrier.
- **Use of Natural Attenuation:** In many cases, natural conditions at contaminated sites meet all the essential environmental factors so that bioremediation can occur without human intervention. This process is called *natural attenuation* and differs from no-action alternatives in that it requires thorough documentation of the role of microorganisms in eliminating the target contaminants. Natural attenuation can be defined as the combination of natural biological, chemical and physical processes that act without human intervention to reduce the mass, toxicity, mobility, volume, or concentration of the

contaminants (e.g., intrinsic bioremediation, dispersion, dilution, sorption, and volatilization). Natural attenuation is typically used in conjunction with active remediation measures (e.g., source removal) or as a follow-up to such measures, and is most appropriate for sites with low concentration of residual fuel hydrocarbons.

1.3.3. How the Presence of Ethanol May Affect Remediation Process Performance

Source removal approaches such as free phase recovery may benefit from changes in the distribution of gasoline when ethanol is present. Solvent flushing techniques using ethanol have been demonstrated to mobilize trapped non-aqueous phase liquids toward recovery wells. Soil vapor extraction will likely have little value in removing ethanol because the ethanol will be strongly partitioned into any unsaturated zone pore water. On the other hand, the increased flow of air through the unsaturated zone will simulate the aerobic biodegradation of ethanol.

The efficiency of pumping systems designed to contain groundwater contaminant plumes should not be directly affected by concentrations of ethanol. However, surface treatment of this water by activated carbon may be adversely affected because the sorption capacity of the filter would be exhausted faster due to ethanol adsorption.

Ethanol is likely to have a negative effect on many current BTEX bioremediation practices. Engineered BTEX bioremediation systems often involve stimulating microbial activities by supplying nutrients and electron acceptors (e.g., O_2 and sometimes NO_3^-), with success often limited by the ability to distribute the stimulating materials throughout the contaminated zone. Ethanol is likely to be present at much higher concentrations than BTEX, which would significantly exacerbate the biochemical oxygen demand (BOD) and nutrient requirements. Thus, maintaining aerobic conditions and precluding nutrient limitation in the contaminated zone could represent a significant increase in treatment costs and a major technical challenge at some sites. In addition, engineered bioremediation works best for high-permeability aquifers ($> 10^{-5}$ m/s), and an ethanol-supported increase in biomass concentration could reduce the permeability of the aquifer and the ability to distribute nutrients and electron acceptors throughout the contaminated zone. Clogging problems near injection well screens and infiltration galleries may also be exacerbated due to additional bacterial growth on ethanol.

In sanitary engineering, anaerobic processes are generally used for pre-treatment of high-strength industrial wastewater (e.g., $BOD > 1,000$ mg/L), because of the high costs and technical difficulties associated with oxygen supply. The effluent from anaerobic reactors is then usually treated aerobically prior to discharge. This suggests that a sequential anaerobic-aerobic approach might be desirable for bioremediation of gasohol-contaminated sites. For example, anaerobic electron acceptors such as nitrate could be injected to accelerate the removal of ethanol and some BTEX contamination. This would alleviate the BOD of the system for more efficient degradation of any residual BTEX in a subsequent aerobic (polishing) stage. Note that anaerobic degradation of ethanol would result in less biomass accumulation (and related clogging problems) than would occur under aerobic conditions, because anaerobic cell yield coefficients are significantly lower. Recent studies have also suggested that anaerobic strategies for the *in situ* bioremediation of petroleum-contaminated subsurface environments may be as effective as aerobic approaches. This notion is similarly based on the fact that introducing sufficient oxygen can be technically difficult and expensive, whereas anaerobic electron acceptors can be easily

added to the subsurface and are chemically more stable (Cunningham *et al.*, 2001; Finneran and Lovley, 2001). Thus, enhanced anaerobic remediation strategies may become more frequently applied to deal with gasohol releases.

As discussed in this report, natural attenuation is likely to be hindered by ethanol, due to its preferential degradation and the accompanying depletion of oxygen and other electron acceptors that would otherwise be available to support BTEX degradation. One possible outcome is that ethanol would increase the distance that BTEX compounds migrate before attenuating processes decrease their concentrations to acceptable levels. Longer BTEX plumes represent a greater probability of exposure to downgradient receptors and thus, decreased acceptability of natural attenuation at some sites.

If a site has an existing MtBE plume, the cleanup of a release of gasohol should not significantly affect the existing site cleanup strategy. Since an MtBE plume can be expected to be more mobile and less biodegradable than either ethanol or BTEX, the monitoring and mass extraction approaches used for MtBE will be generally adequate for ethanol and BTEX as well.

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